Manipulating topology in tailored artificial graphene nanoribbons

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Physical Sciences - Article

Keywords: artificial graphene nanostructures, topological phases, quantum states of matter, topological manipulation

Posted Date: April 15th, 2021

DOI: <https://doi.org/10.21203/rs.3.rs-409243/v1>

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Abstract

Topological phases of matter give rise to exotic physics that can be leveraged for next generation quantum computation^{1–3} and spintronic devices^{4,5}. Thus, the search for topological phases and the quantum states that they exhibit have become the subject of a massive research effort in condensed matter physics. Topologically protected states have been produced in a variety of systems, including artificial lattices^{6–9}, graphene nanoribbons (GNRs)^{10,11} and bismuth bilayers^{12,13}. Despite these advances, the real-time manipulation of individual topological states and their relative coupling, a necessary feature for the realization of topological qubits, remains elusive. Guided by first-principles calculations, we spatially manipulate robust, zero-dimensional topological states by altering the topological invariants of quasi-one-dimensional artificial graphene nanostructures. This is achieved by positioning carbon monoxide molecules on a copper surface to confine its surface state electrons into artificial atoms positioned to emulate the low-energy electronic structure of graphene derivatives. Ultimately, we demonstrate control over the coupling between adjacent topological states that are finely engineered and simulate complex Hamiltonians. Our atomic synthesis gives access to an infinite range of nanoribbon geometries, including those beyond the current reach of synthetic chemistry, and thus provides an ideal platform for the design and study of novel topological and quantum states of matter.

Main Text

Topological insulators (TIs) are materials that belong to an electronic phase of matter that exhibit an insulating bulk coupled with protected and robust in-gap edge states^{14–16}. Their electronic structure cannot be adiabatically deformed into topologically trivial insulators, making them more robust to defects and impurities, and are therefore of great interest to the spintronics and microelectronics communities^{5,17}. In addition, symmetry protected topological (SPT) states are critically important to the emerging field of quantum information science as they are predicted to host Majorana-fermion-like modes when proximity coupled to an s-wave superconductor^{1,2,13}. A promising strategy to explore topological phases is to interface two structures with distinct topological character, consequently triggering the emergence of localized SPT states. This idea was pioneered by Su, Schrieffer and Heeger to describe the in-gap states that exist at domain boundaries in polyacetylene chains¹⁸ and more recently has been exploited to engineer one-dimensional topological bands in graphene nanoribbons (GNRs)^{10,11,19}. In the latter example, SPT states were produced at the junction between armchair GNR segments with varying topological character as defined by their Z_2 invariant – the classification index that differentiates TIs ($Z_2 = 1$) from trivial insulators ($Z_2 = 0$) in gapped one-dimensional systems^{20,21}. The Z_2 invariant of armchair GNRs can be tuned through their width, unit cell and edge termination thus offering a rich landscape for topological band engineering^{19,22,23}. Navigating this landscape requires the ability to build GNRs of arbitrary shape with atomic scale precision on a one-atom-at-a-time basis, an ability that is generally not offered by bottom-up synthesis.

An alternative approach to exploring the band topology of graphene nanoribbons is through the platform of artificial lattices⁷⁻⁹. First demonstrated by Gomes, et al., artificial lattices of bulk graphene are created through the hexagonal arrangement of CO molecules on the surface of Cu (111)²⁴. This is achieved when individual CO molecules are pushed along the surface with a scanning tunneling microscopy (STM) tip to build arbitrary honeycomb structures one artificial atom at a time (Fig. 1a)²⁵. The resulting molecular array repels the surface state electrons into a graphene-like honeycomb lattice, which demonstrates an analogous low-energy electronic dispersion as graphene yet with a reduced Fermi velocity.

In this work, we investigate the emergence and interplay between SPT states in topologically non-trivial artificial GNRs through the real-time manipulation of their geometries and consequently their Z_2 invariants. Additionally, we control the coupling between adjacent states by adjusting their separation and show how it can be used to simulate complex Hamiltonians and quantum states.

Tailored artificial graphene nanostructures are constructed by first using first-principles calculations to identify a GNR which exhibits a non-trivial Z_2 invariant. The Z_2 invariant is determined from the origin independent part of the Zak phase as $Z_2 = \frac{1}{\pi} \int_{-\pi/d}^{\pi/d} dk \langle \psi_{n,k} | \partial_k | \psi_{n,k} \rangle \text{mod} 2$ (Supplementary Information). After identifying the topologically non-trivial nanoribbon geometries, corresponding atomic positions of CO molecules are mapped out and atomically manipulated on the Cu (111) surface such that the space in-between molecules reflect the GNR structure, as illustrated in Fig. 1b. These structures differ from artificial graphene in an analogous way to how GNRs differ from pristine graphene, namely that they are laterally confined. A unique “stitching” was developed for confinement at the edges of our artificial structures.

The bulk-boundary correspondence dictates that boundary modes arise at the interface between a TI and trivial insulator, namely where there is a change in the Z_2 invariant²⁶. We demonstrate this effect at the edge of a topologically non-trivial $N = 9$ armchair GNR ($Z_2 = 1$) and vacuum ($Z_2 = 0$) (Fig. 2a). The configuration of the atomic sites at the termination uniquely determines the nanoribbon unit cell, the origin of which determines the Z_2 invariant in one-dimension¹⁹. In Fig. 2b, we demonstrate the switch between a topologically non-trivial and trivial nanoribbon by constructing an identical 9GNR but removing three artificial atoms from one zig-zag edge and adding them to the other side (Fig. 2b). The origin independent part of the Zak phase of the modified unit cell in Fig. 2b (γ_2') is related to the original unit cell (γ_2) as $\gamma_2' = \gamma_2 - M\pi$, where M is the number of new (or old) lattice sites added (or removed)²³ (Supplementary Information). The topological class of the nanoribbon is changed from $Z_2 = 1$ to $Z_2 = 0$ since we move an odd number of sites effectively homogenizing its Z_2 invariant with the vacuum. To visualize the emergence of boundary modes along the non-trivial interface, we acquire simultaneous STM topographies and normalized differential conductance maps at -55meV of the topologically non-trivial (Fig. 2c) and trivial (Fig. 2d) nanoribbon. The SPT states that decorate the zig-zag edge of the non-trivial nanoribbon vanish when the structure becomes topologically trivial, corroborated by the local density of states (LDOS) (bottom right panels). The LDOS at the zig-zag edge

atoms are further investigated by acquiring dI/dV point spectroscopy (Fig. 2e). The spectrum on the non-trivial edge shows a resonance around -55meV that is not present on the trivial structure suggesting that it emerges from the spatial modulation of its topological character in agreement with the TB LDOS.

To distinguish topology from other mechanisms that could produce edge states we replace the vacuum with a 7GNR assembly. The resulting 7GNR/9GNR heterostructure contains artificial atoms along the interface with the same coordination as those in the bulk. The termination of the 7GNR segment, and hence the choice of unit cell, depends on whether the heterostructure is symmetric (Fig. 3a) or asymmetric (Fig. 3b). The topological class of the symmetric and asymmetric unit cells is $Z_2 = 0$ and $Z_2 = 1$, respectively (Supplementary Information). These results are in agreement with Ref¹⁹ and illustrate the relationship between band topology and nanoribbon unit-cell. To visualize the interface states, STM topographies and normalized differential conductance maps were acquired at -30meV over the artificial atoms of the non-trivial (Fig. 3c) and trivial (Fig. 3d) interface. In agreement with the calculated LDOS maps (bottom panels), the conductance of the two nanoribbons is similar everywhere except for the interface. The dichotomy between the two interfaces is further exemplified in the magnified conductance maps of the non-trivial (Fig. 3e) and trivial (Fig. 3f) interfaces. Line profiles extracted from these maps over the first two rows of interfacial artificial atoms show the states present over the interface where the Z_2 invariant changes that vanish when both sides of the interface are topologically equivalent (Fig. 3g). Furthermore, the dI/dV point spectroscopy acquired on the artificial atom at the center of the interface in Fig. 3e shows a resonance around -30meV that is absent in the spectrum acquired over the analogous atom in Fig. 3f (Fig. 3f), in good agreement with the TB model.

Having established the presence of SPT states that arise from topological inequivalence, we demonstrate how these states and their couplings can be finely engineered to simulate complex Hamiltonians and quantum states. We consider a 9GNR/7GNR/9GNR heterostructure and examine the coupling between the SPT states by varying the lengths of its individual segments (Fig. 4a). This configuration results in 4 topological states ($\psi_{\{A,B,C,D\}}$) at the four interfaces between: (i) vacuum ($Z_2 = 0$) and 9GNR ($Z_2 = 1$), (ii) 9GNR ($Z_2 = 1$) and 7GNR ($Z_2 = 0$), (iii) 7GNR ($Z_2 = 0$) and 9GNR ($Z_2 = 1$) and (iv) 9GNR ($Z_2 = 1$) and vacuum ($Z_2 = 0$). The coupling between the adjacent states depends on their separation ($L_{\{AB,BC,CD\}}$). The TB wavefunctions of the 4 SPT states when they are well separated ($L \rightarrow \infty$) is shown in Fig. 4a.

To examine the range of coupling achievable between adjacent topological states and their robustness, we reduce the length of the middle segment L_{BC} separating B and C while increasing the length of the end segment L_{CD} separating C and D, thereby maintaining a constant total length (Fig. 4b). By moving state C towards a fixed state B and acquiring a line profile over the central resonance of the latter (Fig. 4c), the increased coupling between the two interface states leads to a decrease in the intensity of the peaks at both interfaces. As intensity is proportional to the square of the local components of the eigenstates, such a decrease in intensity demonstrates increased delocalization and the tunability of the wavefunction of the eigenstates resulting from the SPTs. Notably, the coupling between the interface state and end state results in a qualitatively distinct effect with measurable differences between the state

B and C only occurring when $L_{CD} < 3$ unit cells (Fig. 4d). These trends are captured by the TB model (right panel in Fig. 4b and dashed lines in Fig. 4c, d).

The interplay between the four states and the effect of the different segment lengths can be understood by deriving a low-energy 4-state Hamiltonian in the form of a 1D chain with nearest neighbor interactions. As shown in supplementary information, the energy and hopping parameters are determined from TB modeling of the 9GNR/7GNR/9GNR geometry using projectors of states A,B,C,D derived from isolated interfaces and end state calculations. Remarkably, the wavefunctions associated with interface states B and C (end state A and D) are symmetric (anti-symmetric) with respect to the center of the ribbon, yielding , resulting in the low-energy behavior independent of L_{AB} and L_{CD} observed experimentally. The resulting behavior of the eigenstates of the low-energy Hamiltonian thus depends only on the strength of the interactions between interface states B and C. TB parametrization of the hopping terms yields a coupling of 15.6 meV for a one-unit-cell separation, decaying exponentially with separation at a decay constant of 0.524/unit-cell, which reproduces the behavior observed in experiments (Fig. 4) (Supplementary Information).

Our results offer a viable platform to explore and manipulate topological states in GNRs with atomic scale precision over their coupling. Ultimately, the interplay between adjacent SPT states demonstrated here provides fundamental insight into operations requiring the interaction between topological states, such as the braiding of Majorana pairs for topological quantum computation.

Declarations

Acknowledgments: This work was performed at the Center for Nanoscale Materials, a U.S. Department of Energy Office of Science User Facility, and supported by the U.S. Department of Energy, Office of Science, under Contract No. DE-AC02-06CH11357. This material is based upon work supported by Laboratory Directed Research and Development (LDRD) funding from Argonne National Laboratory, provided by the Director, Office of Science, of the U.S. Department of Energy under Contract No. DE-AC02-06CH11357.

Author contributions: DJT, SS, PD, and NPG led the conception, design, acquisition, and analysis of this research along with drafting this manuscript. BLF and CRP provided logistical support for data acquisition. YZ contributed in atomic manipulation and spectroscopy in data acquisition. SWH provided critical guidance and support.

Competing interests: Authors declare no competing interests.

Supplementary Information is available for this paper.

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Figures

Figure 1

Construction of artificial graphene nanoribbons. a, Illustration of the atomic manipulation process used to move CO molecules on a Cu (111) surface. b, (left panel) Identification of a non-trivial nanoribbon, (middle panel) mapping the corresponding CO positions on Cu (111) to obtain the analogous structure and (right panel) STM topography of the realized artificial GNR with $d_{\text{CO-CO}} = 1.0 \text{ nm}$ ($I_{\text{set}} = 0.5 \text{ nA}$, $V_{\text{set}} = 750 \text{ mV}$).

Figure 2

Emergence of topological edge states in $N = 9$ artificial GNR. a, Schematic of the topologically non-trivial $N = 9$ GNR where the red dashed box encompasses the unit cell (inset). b, Schematic of the trivial nanoribbon obtained by moving the three atoms highlighted in red in Fig.2a to the other edge of the nanoribbon. c and d, (top panel) Magnified normalized differential conductance map acquired at -55 meV taken on the zig-zag edge of the non-trivial and trivial nanoribbon, respectively. (bottom-left panel) STM topography, (bottom-middle panel) simultaneously acquired normalized differential conductance map and (bottom-right panel) calculated LDOS map of non-trivial and trivial structure, respectively ($I_{\text{set}} = 1.0$ nA, $V_{\text{set}} = -55$ mV, $V_{\text{mod}} = 20$ mV). $d_{\text{CO-CO}} = 2.0$ nm and scale bars represent 2 nm. e, dI/dV point spectroscopy taken on the positions marked on the STM topographies in Fig.2c and 2d ($I_{\text{set}} = 0.5$ nA, $V_{\text{set}} = -500$ mV, $V_{\text{mod}} = 20$ mV).

Figure 3

Topological interface states in 7/9 heterostructures. a and b, Illustrations of the topologically non-trivial and trivial heterostructure interfaces, respectively. c and d, (top panel) STM topographies, (center panel) normalized differential conductance maps and (bottom panel) calculated TB LDOS maps of the artificial GNRs emulating the schematics from Fig.3a and 3b, respectively ($I_{\text{set}} = 1.0$ nA, $V_{\text{set}} = -30$ mV, $V_{\text{mod}} = 20$ mV). Scale bars represent 2nm. e and f, Magnification of the interfaces outlined by the dashed boxes in Fig.3c and 3d, respectively. g, Cross sectional line cuts of the differential conductance maps along the dashed lines in Fig.3e and 3f. h, dI/dV point spectroscopy acquired on the horizontal indicators in Fig.3e and 3f ($I_{\text{set}} = 0.5$ nA, $V_{\text{set}} = -500$ mV, $V_{\text{mod}} = 20$ mV).

Figure 4

Interplay between SPT states. a, Wavefunctions of 4 SPT states in a 9GNR/7GNR/9GNR heterostructure. The distances between the states are represented as L_{AB} , L_{BC} and L_{CD} , respectively. b, A series of (left column) STM topographies, (middle column) normalized differential conductance maps and (right column) TB LDOS maps of an artificial GNR while varying the separation between states ($I_{set} = 1.0\text{nA}$, $V_{set} = -30\text{mV}$, $V_{mod} = 20\text{mV}$). c and d, Line profiles taken over the central states at site Fig.4b and 4c, respectively, extracted from the conductance maps (solid lines) and the TB LDOS maps (dashed lines) in Fig.4b.

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